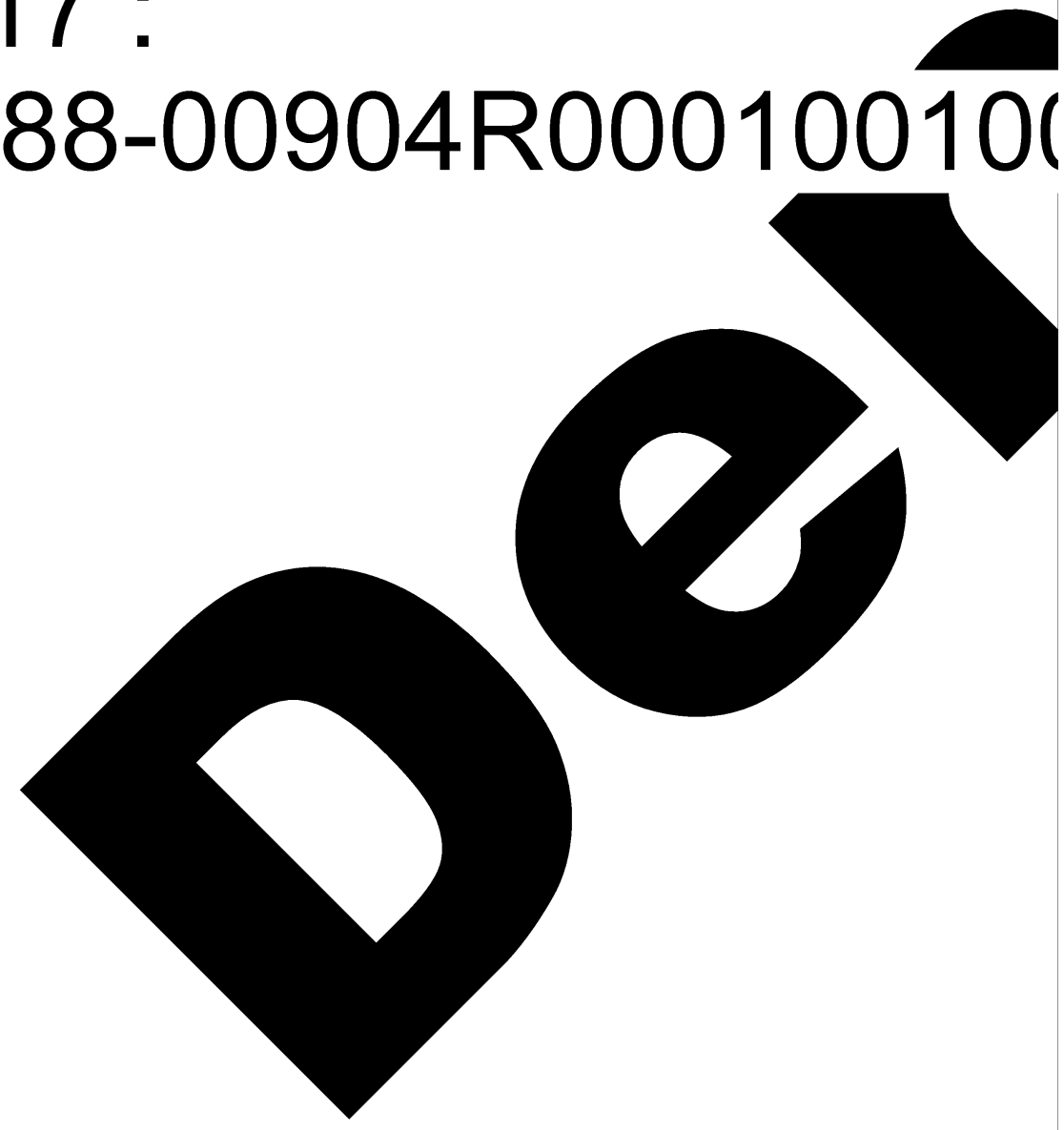

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THE CRITICAL PARAMETERS OF AQUEOUS SOLUTIONS OF $UO_2(NO_3)_2$ AND NUCLEAR SAFETY.

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1. The critical parameters of aqueous solutions of $UO_2(NO_3)_2$.

Critical experiments were performed in reflected and unreflected right circular cylinders, parallelepipeds with square cross section and spheres using an aqueous solution of $UO_2(NO_3)_2$.

The assembly vessels were made from grade 1C18N9T stainless steel with wall thickness of 0.1 - 0.15cm for spheres and cylinders and 0.3cm for parallelepipeds.

In unreflected experiments minimum distance from core to room concrete walls was about 2m.

The solution temperature was changed within the limits of 15 - 21°C, and during a particular experiment the variation was approximately ±0.5°C. Aqueous solutions of $UO_2(NO_3)_2$ contain nitric acid. The value of atomic ratio $\beta_N = \frac{\rho_N}{\rho_U}$ in solutions in experiments with all enrichments and concentrations of uranium was changed within the limits of 2.4 - 3.2. The changes of critical parameters of the solution of $UO_2(NO_3)_2$ enriched to 90% in U^{235} due to the variation of the solution temperature did not exceed errors caused by uncertainty of measurement of the solution volumes and the uranium concentrations. This uncertainty does not exceed ±0.5%.

The experimental data for spheres and parallelepipeds were used to obtain the extrapolation distance and the wa-

ter reflector savings as functions of the uranium concentration and the assembly shape. The critical dimensions for bare parallelepipeds are presented in Table I. The root-mean-square values of geometric bucklings and extrapolation distances for parallelepipeds were obtained from the experiments by the least square method. The errors in the geometric bucklings do not exceed ± 2%. If stainless steel reflector has a thickness T, its saving δ_{ss} satisfies the experimentally found dependence:

$$\delta_{ss} = 0.9\ T,$$

which is valid for $T \leq 1$cm at all assembly shapes in the investigated range of concentrations. The extrapolation distance for parallelepipeds d_{par} does not depend, within the limits of the experimental errors, on uranium concentration in investigated region 72 - 233 g/l:

$$d_{par} = 2.65 \pm 0.15\text{cm}$$

The results of experiments with unreflected spheres and spheres with infinite water reflector are presented in Table II. The extrapolation distance for a sphere d_s is given by

$$d_s = \frac{\pi}{\bar{B}_{par}} - R_o$$

where $\bar{B}^2_{par}$ - buckling deduced from the measurements on bare parallelepipeds; R_o - critical radius of the bare sphere. The extrapolation distance for a sphere in concentration region 72 - 233 g/l does not depend on the uranium concentration and the surface curvature:

$$d_s = 2.7 \pm 0.15\text{cm}$$

The reflector saving due to a complex reflector from stainless steel and water of infinite thickness was measured in special experiments on the parallelepiped, in which the stainless steel thickness varied from 0.3 to 5cm. The reflector saving due to an infinite water reflector for the parallelepiped δ_{par} is a constant, within the limits of the experimental errors, for uranium concentrations from 48g/l to 150g/l:

$$\delta_{par} = 3.4 \pm 0.3\text{cm}$$

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Table I.

Critical data and derived values of $\overline{B^2}$ and $\bar{d}_{par}$ for bare parallelepipeds with square cross section containing solution of $UO_2(NO_3)_2$ (90% enrichment).

Cross section dimension (cm)	U^{235} Density (g/l)	Atomic Ratio ρ_H/ρ_5	Atomic Ratio ρ_N/ρ_U	Critical Height (cm)	$\overline{B^2}$ (cm^{-2})	$\bar{d}_{par}$ (cm)
26.66	73.2	375.1	3.12	46.18		
28.45	73.2	375.1	3.12	36.71		
30.08	73.2	375.1	3.12	31.73	0.0222	2.71
32.63	73.2	375.1	3.12	27.4		
35.55	73.2	375.1	3.12	24.5		
26.66	152.3	175.7	3.08	29.01		
28.45	152.3	175.7	3.08	25.67		
30.08	152.3	175.7	3.08	23.59	0.02679	2.63
32.63	152.3	175.7	3.08	21.39		
35.55	152.3	175.7	3.08	19.76		
26.66	233.4	109.4	3.13	27.08		
28.45	233.4	109.4	3.13	24.20		
30.08	233.4	109.4	3.13	22.32	0.0279	2.67
32.63	233.4	109.4	3.13	20.39		
35.55	233.4	109.4	3.13	18.95		

The reflector saving due to an infinite water reflector for the spheres

$$\delta_s = 3.4 \pm 0.3\text{cm}$$

for uranium concentration from 40 to 230g/l.

The basic experimental results are shown in Fig.1.

The experiments with solution of $UO_2(NO_3)_2$ of 5% and 10% in U^{235} were performed on reflected and unreflected cylinders (see Table III). The geometric bucklings for solutions of low enrichment were obtained by using the extrapolation distance and the water reflector saving from the experiments with the solution of $UO_2(NO_3)_2$ of 90% enrichment. In the present case an error of, say, 8% in $\bar{d}$ could imply an error of 1% in the geometric buckling. The buckling values presented in Table III were computed with an accuracy

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Table II.

Critical Radii for unreflected and reflected spheres containing solutions of $UO_2(NO_3)_2$ (90% enrichment).

Unreflected				Water reflected			
Radius (cm).	U^{235} Density (g/l).	Atomic Ratio ρ_H/ρ_5	Atomic Ratio ρ_N/ρ_U	Radius (cm).	U^{235} Density (g/l).	Atomic Ratio ρ_H/ρ_5	Atomic Ratio ρ_N/ρ_U
20.1	43.2	600	2.5	20.1	32.4	800	2.5
19.0	50.6	500	2.5	19.0	35.5	720	2.5
18.1	67.0	380	2.5	18.1	37.5	680	2.5
17.17	92.8	264	3.3	16.4	50	500	2.5
16.4	112.3	208	2.5	16.0	54	465	2.5
16.14	151.5	156	3.2	13.5	106.4	233	2.5
16.0	194	119	3.15	12.7	172.8	139	2.5
16.0	202.5	118	2.5	12.7	182.0	127.5	3.28
16.0	287	86	2.5				

Table III.

Critical data and values of B^2 for Cylinders Containing solutions of $UO_2(NO_3)_2$ (5% and 10% enrichments).

Enrichment U^{235} (%).	U^{235} density (g/l).	Atomic Ratio ρ_N/ρ_U	Unreflected				Water reflected			
			Diameter (cm)	Critical Height (cm)	d (cm)	B^2 (cm^{-2})	Diameter (cm)	Critical Height (cm)	Effective saving (cm)	B^2 (cm^{-2})
10	48.2	2.5	50	37.4	2.6	0.0128	40 x)	36.8	6.0	0.0128
10	42.2	3.1	50	39.7	2.6	0.0123	50 x)	31.7	6.0	0.0122
10	32.2	3.0	50	54.1	2.6	0.0103	50 x)	40.7	6.0	0.0101
10	25.0	3.0	60	60.2	2.6	0.0^2767	50 x)	70.8	6.0	0.0^2763
5	25.3	2.4	80	70.4	2.6	0.0^2487	72.5	65.7	6.0	0.0^249

x) No top reflector.

of 2%. By setting the geometrical buckling in systems having different geometries but the same composition to be equal, radii of reflected and unreflected spheres were obtained from cylinder data of Table III (see Fig.2).

In present report the P_1 and P_3 approximations of the multigroup version of the spherical harmonics method have been used for a solution of the Boltzmann's transport equation [1],[2]. The calculations have been performed on the electronic computers. When calculating reactors in the P_1 approximation, the scattering in the slowing-down region on all nuclei except hydrogen was considered in the age-diffusion approximation. The scattering on hydrogen was calculated exactly. The multigroup constants were calculated from the basic cross section data [3],[4],[5]. The resonance absorption was considered in the approximation of the narrow resonances. The Doppler broadening of neutron resonances was calculated according to Ref.[6]. The interference between resonance and potential scattering processes was neglected. The effective one-group constants in the thermalization region were found by means of averaging over the neutron spectrum satisfying Wilkins' equation[7]. The chemical binding of hydrogen in the water molecule is taken into account by the effective mass equal about 6. The system of constants for hydrogen and oxygen was tested by comparison of theoretical and experimental values of the square of slowing-down length (L_s^2) to indium resonance energy and the diffusion length of thermal neutrons (L) (Table IV).

Table IV.

Value	Source	Experiment [8]	Experiment [9]	Experiment [10]	Calculation
L_s^2		27.1 0.9	27.86 0.10		27.8
L				2.72	2.6

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To compare theory and experiment on the critical masses in the wide region of concentrations it is necessary to perform all calculations in the P_3 approximations. The reactor critical dimensions can also be found with sufficient accuracy by means of the effective one-group theory [11] taking into account the transport effects. The calculation in the multigroup P_3 approximation of the critical radius of the water reflected sphere containing solution of $UO_2(NO_3)_2$ (90% enrichment) coincides with experiment within $\pm$1.5%. The calculation of critical radius of the unreflected sphere exceeds the experimental results by 2 - 2.5% (Fig.1). The same conclusions are drawn from the comparison of the experiment and the calculation for solutions of $UO_2(NO_3)_2$ enriched to 10% in U^{235} (Fig.2). The single experiment for the solution of 5% enrichment gives the values of critical radii of the reflected and unreflected spheres exceeding the values obtained by the calculation in the multigroup P_3 approximation by 1%. Thus, the calculation of unreflected reactor in the region of the large uranium concentrations is of a less accuracy than calculations of reflected reactor. The calculation of the reflected sphere critical radius as a function of concentration (90% enrichment) at the various nitrogen to uranium ratioes shows, that the minimum radius of reflected sphere increases by 7 - 8%, when atomic ratio ρ_N/ρ_U increases from 2 to 6.3. The maximum difference of critical radii calculated by the one- and multigroup P_3 approximations is 2.5 - 3% and increases with the increase of concentration.

The material buckling of the solutions of $UO_2(NO_3)_2$ is calculated by

$$B^2 = \left(\frac{\pi}{R_3 + d_s}\right)^2$$

where R_3 - the critical radius of the bare sphere in the multigroup P_3 approximation; d_s - the extrapolation distance in the P_1 approximation: $d_s = \frac{\pi}{B_1} - R_1$, where B_1^2 - the

material buckling in the P_1 approximation; R_1 - the critical radius of the bare sphere computed in the multigroup P_1 approximation with energy - dependent extrapolation distance. The extrapolation distances for an infinitely long cylinder (d_c) and a slab of infinite area ($d_{s\ell}$) were computed on the assumption that the linear extrapolation distance is independent of the core shape. The dependence of d_s, d_c and $d_{s\ell}$ on the uranium concentration is shown in Fig.1. The extrapolation distance increases with the incresing of concentration and the curvature of the core surface. The difference between the experimental and computed values of the extrapolation distance for the sphere is as much as 7%. The difference in the material buckling for the solutions of 90% enrichment in the concentration region 72 - 233 g/l is as much as 8%. The basic difference (7%) is due to the discrepancy of the computed and experimental values of the critical radii of the bare spheres. The discrepancy in the extrapolation distance of 7% leads to discrepancy in the buckling, which is no more than 1%. For the solution of 5% and 10%-enrichments the discrepancy between the calculation and the experiment is as much as 6%: 1% due to the difference in the extrapolation distances, 5% due to the difference in the radii of bare spheres.

The water reflector saving for sphere is found as difference between the critical unreflected and reflected dimensions. According to the calculation the reflector saving is directly proportional to the uranium concentration (the curve 4 in Fig.1). The discrepancy between the calculation and the experiment is as much as 10%.

The empirical kernel method [12] is one of the methods dealing with criticality problems. In this method the critical condition is obtained from a measured kernel without recourse to any mathematical model to describe the neutron slowing-down process. The empirical kernel method can be applied to bare cores containing a dilute solution of U^{235}

in water if: 1) fission is confined to thermal region; 2) the core dimensions are such, that one can neglect the energetic dependence of the extrapolation distance. The systems containing the solutions with the large concentration of U^{235} do not satisfy these requirements. Unfortunately there are no data concerning the influence of the displacement of water by the molecules of $UO_2(NO_3)_2$ on the neutron slowing-down in the solution. In the present report this problem is considered qualitatively. One can assume [12] that there is no correlation between neutron leakage and neutron absorption. According to the asymptotic theory for a bare uniform reactor the condition for the criticality has the following form [12]:

$$\overline{K}(B;1,4)\, e^{-B^2\Delta\tau_{th}}\left[k_{\infty f}+\frac{k_{\infty th}}{1+B^2L^2}\right]=1 \qquad (1)$$

provided the thermal group is responsible for major part of fission. $\overline{K}(B;1.4)$ is the nonleakage probability for neutrons reaching 1.4ev. $\overline{K}(B;1.4)$ was computed by M.N.Lantsov [13] from the experimental data of Hill, Roberts and Fitch [14] in such a way that the square of the slowing-down length to indium resonance energy in the water is $27.8cm^2$. The quantities $k_{\infty f}$, $k_{\infty th}$ are computed in the multigroup P_1 approximation. The material buckling B^2 found from eq.(1) is presented in Fig.1 (curve 3a). In computing B no account has been taken of the effect of the displacement of water by the molecules of $UO_2(NO_3)_2$. The comparison of the curve 3a and the experimental data for the parallelepipeds shows, that this assumption is not justified. Rewrite (1) as

$$\overline{K}\left(\frac{B}{\rho^{\alpha}};1,4\right) e^{-\frac{B^2\Delta\tau_{th}}{\rho^{2\alpha}}}\left[k_{\infty f}+\frac{k_{\infty th}}{1+B^2L^2}\right]=1 \qquad (1a)$$

where ρ - the effective density of hydrogen in the solution;
α - a factor, taking into account the influence of the molecules of $UO_2(NO_3)_2$ on the nonleakage probability $\overline{K}(B;1.4)$. The comparison with the experimental values of the geometric buckling shows, that "α" decreases with increase of uranium concentration (curves 3b, 3c, 3d, 3e in Fig.1).

This fact indicates, that the importance of scattering collisions with nonhydrogen atoms increases with increase of uranium concentration. The calculation for 90% enrichment shows, that at the same uranium concentration material buckling decreases with atomic ratio ρ_N/ρ_U, 80% being due to decrease of hydrogen density and 20% being due to nitrogen absorption. For uranium solution of lower enrichment the importance of nitrogen absorption increases.

2. The effectiveness of heterogeneous neutron absorbers in aqueous solutions of $UO_2(NO_3)_2$ (10% and 90% enrichments).

Critical experiments with neutron absorbers have been carried out in cylindrical assemblies 40cm and 110cm in diameter. Single cylindrical and circular absorbing rods, a group and a lattice of absorbing rods were inserted into a core of assembly, with contained solution of $UO_2(NO_3)_2$. Two neutron absorbing materials were used: 1) a cadmium sheet of 0.05cm thickness; 2) boron carbide powder of 1.25 g/cm^3 density. The neutron absorbers had the stainless steel sheaths.

The experiments have been carried out under conditions described in section 1. The uncertainty of measurement of the solution critical heights is estimated at no greater than $\pm$ (1.5 - 2%). Assembly of diameter 40cm. The effectiveness of a central cylindrical absorbing rod extending throughout the whole length of a cylindrical core depends on the material of a rod, its diameter and uranium concentration in solution. The absorbing rod is a stainless steel tube which contains the absorbing material. In experiments with the boron carbide rods the sheath thickness was varied (Fig.3). The presence of steel sheath leads to two contrary effects: 1) screening of boron carbide; 2)displacement of solution. It appears from Fig.3 that the displacement effect prevails. The effectiveness of central absorbing rod consisting of boron carbide with steel sheat was computed accor-

ding to one-dimensional reactor code in multigroup P_1 approximation on electronic computer. The effective boundary condition on the outer surface of the absorbing rod depends on the albedo of the rod. The albedo of the canned rod was computed according to the method proposed by E.I.Grishanin [15]. The calculation agrees with the experiment (Fig.3) within 10 - 25%. The agreement improves with the increasing of rod diameter.

The experimental comparison of the effectiveness of central boron carbide rod and central water-moderated cadmium rod is shown in Fig.4. If rod diameter is less than approximately two slowing-down lengths ($2L_s$) boron carbide rod is more effective than water-moderated cadmium rod. With an increase of rod diameter the selfshielding of boron carbide rod decreases its effectiveness, while the number of neutrons slowed-down in the water-moderated cadmium rod below the cadmium cut-off and absorbed by cadmium is increased. If the rod diameter is more than $2L_s$, the boron carbide rod is less effective than the water-moderated cadmium one.

The experiments on a system (a rod in the centre and a ring of equidistantly placed rods) of boron carbide rods (outer diameter 5.8cm, sheath thickness 0.4cm) and water-moderated cadmium rods (outer diameter 5.7cm, sheath thickness 0.55cm) were also carried out. The effectiveness of a system of absorbing rods reaches its maximum (Fig.5) at certain distance between rod centers. This distance depends on the uranium concentration, rod type and reactor reflector.

The effectiveness of annular boron and cadmium absorbers versus its mean radius is shown in Fig.6. The annular gap thickness is 0.6cm, the stainless steel sheath thickness is 0.1cm. As seen from Fig.6, the optimal mean radius of annular absorber in a bare core is about 0.45R, where R - radius of a core. Similar experiments in a reflected reactor give the optimal radius of the absorber equal about 0.53R. The annular absorbers with boron carbide are more ef-

fective than the absorbers with cadmium. When filling with water the annular absorber containing two coaxial cadmium cylinders its effectiveness increases with thickness of water gap (Fig.7).

Assembly of diameter 110cm. In critical experiments on assembly 110cm in diameter the effectiveness of absorbing rods arranged in triangular lattice was found. The control rod is the stainless steel tube (outer diameter 5.5cm, sheath thickness 0.5cm), which contains the boron carbide powder. The positioning and the fixation of absorbing rods in lattice were provided by two steel plates 1.7cm in thickness (one of them placed on the core bottom, the other on the top of the core vessel). The results of the experiments are represented in Table V.

For estimating the effectiveness of the absorbing-rod lattice the core was divided into two homogeneous regions: region occupied by the rods and region free of rods. The absorption in the rods is found from the analysis of regular lattice by the method of an equivalent cell with an effective boundary condition on the absorbing rod surface. In the case of a dense lattice allowance is made for a mutual screening of rods. The absorbing properties of a rod in a cell are characterized by an effective macroscopic cross section $\Sigma_{eff}(u)$ determined by the rod composition, the lattice spacing and the neutron energy. The energy dependence can be obtained by either the group method [16] or the age-diffusion approximation. Let us assume that in the age-diffusion approximation the neutron slowing-down density can be represented by:

$$q(\rho,u) = R_u(\rho)\, e^{-\int_0^u \frac{\Sigma_c}{\xi\Sigma_s}\,du' - \int_0^u \frac{\mathcal{H}^2 D}{\xi\Sigma_s}\,du'}$$

The basic energy dependence is represented by the exponential. The function $R_u(\rho)$, which gives the spatial distribution of the slowing-down density, satisfies equation:

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$$\Delta R_u(\rho) + \mathcal{H}_u^2 R_u(\rho) = 0$$

with appropriate boundary conditions [16]. Computed in the P_1 approximation critical heights of solution in assembly 110cm in diameter with triangular lattice of rods (outer diameter 5.5cm and sheath thickness 0.5cm) are presented in TableV. The calculation of $\Sigma_{eff}(u)$ was carried out in age-diffusion approximation taking into consideration the mutual sereening of absorbing rods in a dense lattice. The dependence of the diffusion and slowing-down properties of the region filled with rods on lattice spacing was not taken into account. It understimates the critical calculated heights. The comparison of calculation and experiment shows, that the age-diffusion method of calculation of $\Sigma_{eff}(u)$ overstimates the rod effectiveness at a dense lattice and understimates the rod effectiveness at widely-spaced lattice.

Table V.

Critical heights of solution of $UO_2(NO_3)_2$ in assembly $\emptyset$ 110cm with side and bottom reflector. The lattices of absorbing rods $\emptyset$5.5 x 0.5cm.

No.	U^{235} Enrichment (%)	Uranium Density (g/l)	Lattice spacing (cm)	Rod Number	Critical Height H (cm) Calculation	Experiment	$H_{calc}-H_{exp}$ / H_{exp} (%)
1	5.64	400.2	31.8	7	-	103.8	-
2	10	420.5	22.8	19	-	27.4	-
3	10	420.5	18.4	31	-	34.2	-
4	90	37.51	13.4	51	-	102	-
5	90	37.51	22.8	19	23.3	23.0	+1.5
6	90	37.51	15.2	41	38.8	40	- 3
7	90	37.51	7.6	41	26.1	24.7	+ 6
8	90	74.87	21.2	19	15.6	17.0	- 8
9	90	74.87	15.2	40	20.3	20.0	+ 1.5
10	90	74.87	13.2	51	23.4	24.2	- 3.0
11 x)	90	152.3	21.2	19	13.2	13.3	+1.0
12 x)	90	152.3	7.6	61	14.6	14.5	+1.0

x) Without plate on core bottom.

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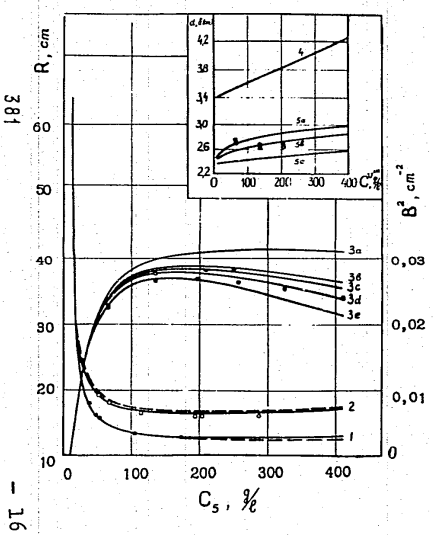

Fig.1. Critical parameters of $U(90)O_2(NO_3)_2 - H_2O$ solutions. Ratio $\frac{\rho_N}{\rho_U}$ = 2.5. Radius of sphere R:1 - reflected; 2 - bare; ○, ⊗ - experiment; —— - P_3 multigroup computation; ——P_3 one - group computation.
Material buckling B^2: 3 - by Kernel method, a - from $\bar{K}(B)$([13]), b - $\bar{K}(\frac{B}{\rho^{0,5}})$, c - $\bar{K}(\frac{B}{\rho^{0,6}})$, d - $\bar{K}(\frac{B}{\rho^{0,75}})$, e - $\bar{K}(\frac{B}{\rho})$; ⊙ - experiment, ⊕ - computation by $B^2 = \left[\frac{\pi}{R_3+d_s}\right]^2$. 4 - reflector saving δ; 5 - extrapolation distance d: a - for sphere; b - for infinitely long cylinder; c - for slab of infinite area; experiment: ● - sphere data; ⊡ - parallelepiped data.

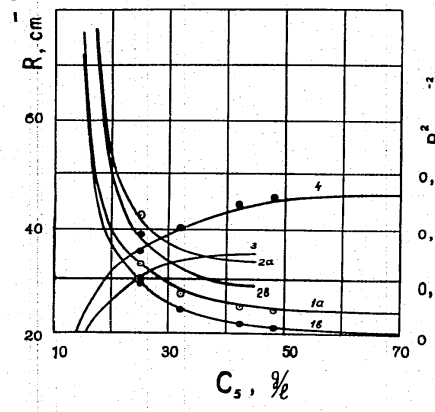

Fig.2. Critical parameters of $UO_2(NO_3)_2 - H_2O$ solutions. Ratio $\frac{\rho_N}{\rho_U}$ = 2.5. Radius of sphere R in P_3 multigroup computation: 1 - 10%, 2 - 5% enrichments; a - bare, b - water reflected.
Material buckling $B^2 = \left[\frac{\pi}{R_3+d_s}\right]^2$: 3 - 5%, 4 - 10% enrichments. ○, ⊗, ⊕ - experiment (from cylinder data).

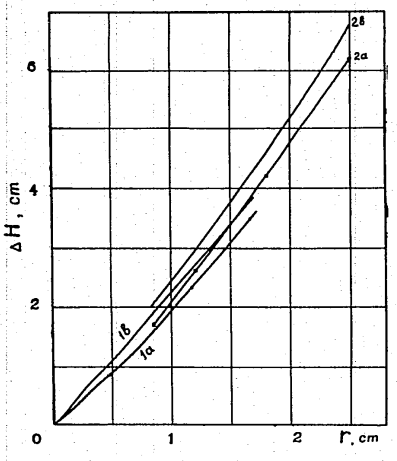

Fig.3. Critical height increment as a function of absorbing rod radius (on boron carbide). Bare assembly of 40 cm diameter. Uranium concentration 72 g/l (90% enrichment).
1 - steel sheath 0.03 - 0.05 cm in thickness; 2 - sheath 0.4cm in thickness.
a - experiment;
b - computation.

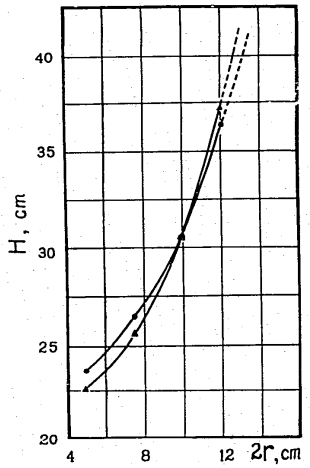

Fig.4. Critical height as a function of absorbing rod diameter. Bare assembly ϕ 40cm. Uranium concentration 136 g/l. Steel sheath 0.1cm in thickness.
▲ - water-moderated cadmium rod;
● - boron carbide rod.

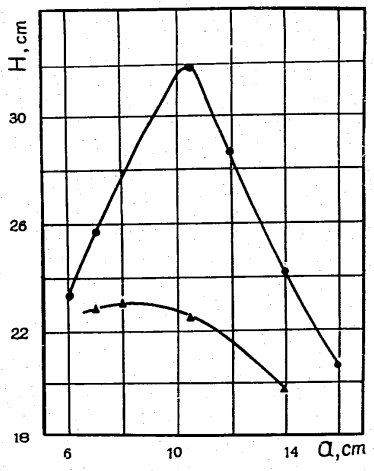

Fig.5. Effectiveness of seven absorbing rods as a function of distance a between their centers. Assembly of 40cm in diameter with side and bottom reflector (90% enrichment).
● - boron carbide rods. Steel tube 5.8 x 0.4cm in diameter.
C_U = 289 g/l.
▲ - water moderated cadmium rods. Steel tube 5.7 x 0.55cm in diameter contains cadmium tube ϕ4.6 x 0.05cm.
C_U = 457 g/l.

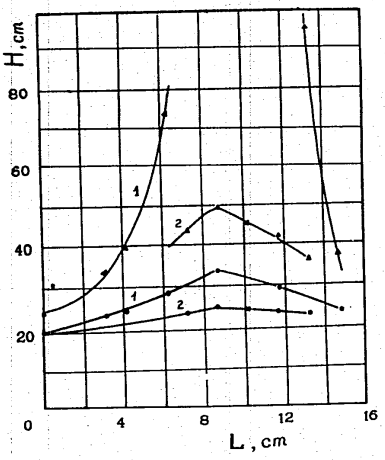

Fig.6. Critical height as a function of mean radius of annular absorber L.
Bare assembly 40cm in diameter. Steel sheath of absorber 0.1cm in thickness (90% enrichment).
1. Boron carbide absorber. Annular gap thickness 0.6cm. 2. Cadmium absorber. Cadmium thickness 0.05cm.
▲ - uranium concentration 76 g/l;
◐ - uranium concentration 289 g/l.

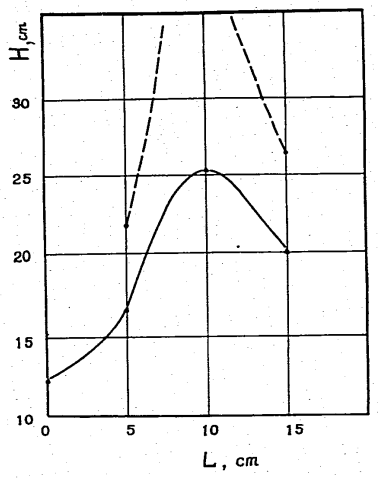

Fig.7. Critical height as a function of mean radius of annular water - moderated absorber L. Assembly ∅ 40cm with side and bottom water reflector. Steel sheath 0.1cm in thickness. Uranium concentration 289 g/l (90% enrichment).
Distance between two coaxial cadmium cylinders: ▲ - 2.5cm;
● - 5 cm.